

Revisiting the evidence of increasing springtime ozone mixing ratios in the free troposphere over western North America

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Draft: October 5, 2015

Submitted to *Geophysical Research Letters*

GRL length limit (3 figures + ~4500 words)

Key Points:

- (1) Multi-decadal hindcast simulations to interpret O₃ trends based on incomplete observations
- (2) Large variability in meteorology and sparse *in-situ* sampling complicates O₃ trend estimates
- (3) While rising Asian emissions raise U.S. O₃ 'background', the model 'true median' indicates weaker increases

Abstract

We present a 20-year time series of in-situ free tropospheric ozone observations above western North America during springtime and interpret results using the GFDL-AM3 chemistry-climate model hindcast simulations (1980-2014). Revisiting the analysis of *Cooper et al.* [2010], we show that sampling biases can substantially influence calculated trends. AM3 co-sampled in This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1002/2015GL065311

space and time with observations reproduces the observed ozone trend (0.65 ± 0.32 ppbv year⁻¹) over 1995-2008 (in simulations either with or without time-varying emissions), whereas AM3 ‘true median’ with continuous temporal and spatial sampling indicates an insignificant trend (0.25 ± 0.32 ppbv year⁻¹). Extending this analysis to 1995-2014, we find a weaker ozone trend of 0.31 ± 0.21 ppbv year⁻¹ from observations and 0.36 ± 0.18 ppbv year⁻¹ from AM3 ‘true median’. Rising Asian emissions and global methane contribute to this increase. While interannual variability complicates the attribution of ozone trends, multi-decadal hindcasts can aid in the estimation of robust confidence limits for trends based on sparse observational records.

1. Introduction

The lifetime of ozone in the free troposphere is on the order of several weeks, sufficiently long for ozone to be affected by changes in hemispheric precursor emissions and large-scale atmospheric circulation patterns. Since 1990, anthropogenic emissions of ozone precursors have tended to shift from North America and Europe to Asia [Richter *et al.*, 2005; Granier *et al.*, 2011]. This rapid shift has motivated international cooperative efforts to gather the best available measurements and to assess the global and regional ozone distribution and changes [e.g. Logan *et al.*, 2012; Parrish *et al.*, 2012; Oltmans *et al.*, 2013]. Cooper *et al.* [2010] compiled mid-tropospheric ozone measurements from all available suborbital platforms across western North America and found a significant springtime ozone increase of ~ 0.6 ppbv year⁻¹ during 1984-2008. This ozone growth rate is much higher than simulated by free-running chemistry-climate models with historical emissions [Lamarque *et al.*, 2010; Parrish *et al.*, 2014]. During springtime, meteorological variability in ozone over western North America is large and heterogeneous in space and time [Lin *et al.*, 2015]; as such, changes in observing practice or small sample size can

complicate efforts to obtain robust estimates of ozone trends. As an example, we revisit the extensive analysis of *Cooper et al.* [2010] and interpret the representativeness of observation records using chemistry-climate model hindcasts [*Lin et al.*, 2014].

Analysis of available observations indicates an increase ($0.3\text{--}0.8\text{ ppbv year}^{-1}$) in springtime ozone at some rural western U.S. sites during the 1990s and the 2000s [*Jacob et al.*, 1999; *Jaffe and Ray*, 2007; *Parrish et al.*, 2009; *Cooper et al.*, 2012; *Fine et al.*, 2014; *Gratz et al.*, 2014]. The ozone trends inferred from these 10–20 years of observations cannot be reproduced by increasing Asian emissions and global methane in chemical transport model experiments with single-year meteorology [*Zhang et al.*, 2008; *Fiore et al.*, 2009]. *Parrish et al.* [2014] find that current free-running chemistry-climate models capture only ~50% of ozone changes observed over recent decades, raising concerns about missing processes in the models. However, we note that such free-running models are not expected to fully represent the influence of circulation variability on long-term ozone measurements. Interannual to decadal variability in circulation regimes (i.e., internal climate “noise”) can confound the attribution of observed ozone changes to anthropogenic emission trends, as found in the 40-year ozone record at Mauna Loa Observatory in Hawaii [*Lin et al.*, 2014].

A number of studies have emphasized the importance of stratosphere-to-troposphere transport (STT) for interannual variability of tropospheric ozone at northern mid-latitudes [*Ordonez et al.*, 2007; *Cuevas et al.*, 2013; *Neu et al.*, 2014; *Hess et al.*, 2015]. The western U.S. is particularly susceptible to the STT influence. *Lin et al.* [2015] find that stratospheric intrusions can explain much of the year-to-year variability of springtime surface ozone means and extremes

measured at western U.S. mountain sites during 1990-2012. In light of large meteorological variability, sampling deficiencies can substantially influence calculated ozone means and trends, as illustrated for a mountain site [*Fischer et al.*, 2011], ozonesondes [*Saunio et al.*, 2012; *Thompson et al.*, 2014; *Lin et al.*, 2015], and satellite instruments [*Toohey et al.*, 2013].

We build on earlier work by systematically evaluating changes in springtime ozone levels above western North America using a suite of 35-year hindcast simulations (1980-2014) conducted with the Geophysical Fluid Dynamics Laboratory global chemistry-climate model (GFDL-AM3) nudged to reanalysis winds. These hindcast simulations (as opposed to free-running climate models) enable us to develop an apples-to-apples comparison, both in space and time, between observations and model simulations. We seek to answer the following questions: (1) How representative are the ozone trends derived from the current measurements with sparse spatial and temporal sampling over western North America? (2) Can the model reproduce observed ozone interannual variability and trends? (3) To what extent can changes in springtime ozone above western North America over recent decades be attributed to meteorological variability versus trends in Asian anthropogenic emissions and global methane?

2. Methods

The GFDL-AM3 simulations are designed to isolate the response of ozone to historical changes in anthropogenic emissions and meteorology, as described in *Lin et al.* [2014; 2015]. The BASE simulation applies interannually-varying emissions of aerosol and ozone precursors from human activity, based on *Lamarque et al.* [2010] for 1980-2000 and Representative Concentration Pathway (RCP) 8.5 projections [*Riahi et al.*, 2011] beyond 2005, linearly

interpolated for intermediate years. Global methane abundance under BASE increases by 16% from 1980 to 2014. The FIXEMIS simulation, with anthropogenic emissions set to the 1970-2010 climatology and methane held constant at 2000 levels, is designed to isolate the role of meteorology. All simulations include interactive stratosphere-troposphere chemistry and aerosols at approximately 2° by 2° horizontal resolution [Austin *et al.*, 2013; Naik *et al.*, 2013], with pressure-dependent nudging to the NCEP/NCAR reanalysis winds [Lin *et al.*, 2012a,b]. Despite mean state biases (12%; **Fig.S1**), AM3 captures the salient features of tropospheric ozone over western North America, including the influences from Asian pollution and deep STT events [Lin *et al.*, 2012a,b], as well as their variability on interannual to decadal time scales [Lin *et al.*, 2014; 2015], indicating that the model represents the underlying processes controlling ozone variability, and is thus a suitable tool for studying the representativeness of under-sampled observational records.

The ozone data used by Cooper *et al.* [2010] were assembled from all available ozonesondes, lidar and commercial and research aircraft observations across western North America (25°-55°N, 130°-90°W, 3-8 km altitude) during April-May in years 1984 and 1995-2008. Ozone sample sizes range from 1,826 in 1984 to 8,769 in 2006. Cooper *et al.* [2010] suggested that a minimum sample size of 1200 measurements per April-May period was required for all years. The years between 1985-1994 had too few data, with data availability limited to the weekly ozonesondes from Boulder and Edmonton. As these observations were not made explicitly for trend monitoring purposes, the observing frequency, timing, and locations of measurements change from year to year. Cooper *et al.* [2010] conducted extensive statistical

tests of the robustness of their estimated trends by sub-sampling the datasets in several ways: limiting the latitude range of the data to 25°-45°N, removing all the research aircraft campaign data, focusing on air masses transported directly from Asia, and removing data with recent influence from the North American surface. They concluded that spatial and temporal biases in the observations do not have a major impact on the ozone trend inferred for 1995-2008.

We revisit the robustness and representativeness of the derived trend by comparing the median values of measurement samples and model results co-sampled with available observations in space and time with the ‘true median’ value (i.e., continuous temporal and spatial sampling over western North America) determined from daily ozone fields archived from the model. Trends in the time series data are determined using an ordinary linear least squares fit. We report the slope of the linear fit, the 95% confidence intervals, and p-values for a two-tailed t-test. To facilitate comparison with trends reported in the earlier analysis, we do not adjust the confidence intervals for serial autocorrelation.

3. Representativeness of ozone profile observations in western North America

3.1 The 1984-2008 and 1995-2008 trends

[Figure 1 about here]

The composite of observations reported by *Cooper et al.* [2010] shows an ozone increase of 0.65 ± 0.32 ppbv year⁻¹ ($p < 0.01$) for April-May during 1995-2008 and suggests a similar rate of increase (0.66 ± 0.20 ppbv year⁻¹) since 1984 (Black circles in **Fig.1**). With both emissions and meteorology varying, AM3_BASE co-sampled in space and time with available observations simulates an ozone increase of 0.75 ± 0.38 ppbv year⁻¹ ($p < 0.01$) during 1995-2008 (orange circles

in **Fig.1a**). For comparison, the model ‘true median’, with continuous daily and spatial sampling over western North America, indicates much weaker and insignificant increases at the 95% confidence level: 0.25 ± 0.32 ppbv year⁻¹ ($p=0.12$; red circles in **Fig.1a**). Even with emissions held constant, AM3_FIXEMIS when co-sampled reproduces much of the observed interannual variability ($r^2=0.56$) and simulate ozone increases of 0.68 ± 0.38 ppbv year⁻¹ ($p<0.01$) during 1995-2008 and 0.56 ± 0.25 ppbv year⁻¹ ($p<0.01$) during 1984-2008 (**Fig.1b**). In striking contrast, no significant ozone increase is found in the FIXEMIS ‘true median’: 0.14 ± 0.24 (0.02 ± 0.11) ppbv year⁻¹ for 1995-2008 (1984-2008). While the model co-sampled and ‘true’ median trends have overlapping confidence intervals, they are significantly different at the 95% confidence level based on a t-test (see Supplementary Methods).

The ability of the model to reproduce the observed ozone growth rates when sampled sparsely as in the observations, but not when sampled continuously in space and time, indicates that large meteorological variability and sparse sampling complicate the ozone trend estimate in *Cooper et al.* [2010], despite their combining as many observational datasets as possible and testing for robustness of the trend by sub-sampling. Observations are strongly correlated with BASE when co-sampled ($r^2=0.71$) but only weakly correlated with the BASE ‘true median’ ($r^2=0.45$); the corresponding statistics with FIXEMIS are 0.56 versus 0.21. The largest discrepancies between the model co-sampled and ‘true median’ occur in 1984, 1999, 2001, 2005, 2006, and 2009. The model co-sampled value for 2001 is biased high as compared to observations but they agree well when aircraft campaign data are removed (Fig.1c). Compared to the BASE ‘true’ ozone median, the observed and model co-sampled values are biased low by

~10 ppbv in 1984 but biased high by 7 ppbv in 2005 and 2006. These sampling artifacts augment the apparent rate of ozone increase in the observed (and co-sampled) dataset during 1984-2008 and 1995-2008.

[Figure 2 about here]

The influence of meteorology on interannual and geographical variability of springtime ozone in the free troposphere is much stronger than emission changes. We thus use AM3_FIXEMIS to discuss the influence of sampling deficiencies on observational estimates of regional median ozone (Fig.2). AM3 forced with observed meteorology indicates that the distribution of mid-tropospheric ozone across western North America during spring varies from year to year depending on the location of the mid-latitude jet stream that influences long-range transport of Asian pollution and stratospheric ozone intrusions [Lin *et al.*, 2015]. The model shows a latitudinal ozone gradient of 10-20 ppbv between 40°-55°N and 25°-35°N along the U.S. west coast, consistent with the observed ozone gradient at sonde sites from north to south in California [Cooper *et al.*, 2011]. Flight tracks of the 1984 CITE-1 aircraft campaign [Hipskind *et al.*, 1987] were located between 30° and 40°N, where mean ozone levels are substantially smaller than north of 40°N (**Fig.2a**). During the 2006 INTEx-B campaign, in contrast, the flights were between 40° and 50°N, where mean ozone amounts are much larger (**Fig.2c**). To control for latitudinal sampling bias, Cooper *et al.* [2010] found little change in the median ozone rate of increase when the latitudinal range of observations was restricted from 25°-55°N to 25°-45°N. However, AM3 suggests the spatial scale of meteorological variability in mid-tropospheric ozone between 25°-45°N latitude and across 130°W-90°W longitude is not captured by the available

observations (**Fig.2**) and has large enough amplitude to confound the calculated ozone trend. The observed median ozone is highest in spring 2006 (Fig.1a), attributed in part to INTEX-B sampling of Asian pollution plumes.

The year-to-year meteorological variability is important even if the measurements are made at the same places (++symbols in **Fig.2a vs. Fig.2b**). *Parrish et al.* [2004] compared data from the 1984 CITE-1 and 2002 ITCT-2K2 campaigns for the overlapping time of year and region (30°-40°N latitude and 105°-135°W longitude) and for aged air masses (i.e., eliminating data with ozone levels above 100 ppbv to minimize the effects of stratospheric intrusions). They found a marked change in the photochemical environment of the North Pacific troposphere, with a median ozone increase of ~10 ppbv between the two campaigns. Using the same data selection procedure as in *Parrish et al.* [2004], we find a median ozone increase of ~11 ppbv in AM3_BASE and ~5 ppbv in FIXEMIS, indicating that meteorological variability and emission trends contribute equally to the observed ozone increase between the two campaigns. Unusual transport of tropical air to the southwest U.S. has influenced observations from the 1984 CITE-1 campaign, as supported by the model with constant emissions, which captures the substantially lower ozone observed in spring 1984 than in the later years (**Fig.1b**).

With the research aircraft campaign data removed (**Fig.1c**), *Cooper et al.* [2010] found an ozone increase (0.69 ± 0.29 ppbv year⁻¹) similar to that derived from all available data for 1995-2008. Both BASE and FIXEMIS co-sampled with this more limited dataset capture the observed interannual variability ($r^2 \sim 0.7$) and simulate a significant ozone increase of ~0.7 ppbv year⁻¹, in contrast to the insignificant ‘true median’ trends (**Fig.1c**). Anomalously frequent intrusions of

stratospheric ozone were observed during April-May 1999 [Langford *et al.*, 2009], as a result of the polar jet meandering towards the western U.S. following the extreme La Niña event of 1998-1999 [Lin *et al.*, 2015]. The MOZAIC commercial aircraft profiles and weekly ozonesonde measurements are too infrequent and sparse to precisely characterize the actual high-ozone anomaly in April-May 1999 (**Fig.1c** and **Fig.2d**). For 2005, Trinidad Head ozonesonde, Table Mountain ozone lidar, and MOZAIC profiles over California contribute 77% of the data (**Fig.2e**). Although these measurements do not target particular features, they do sample the enhanced ‘background’ ozone flowing into the southwest U.S. in April-May 2005 compared to other years (see also **Fig.S2**). We conclude that large meteorological variability confounds the ozone trend estimate from sparse observations, regardless of whether the research aircraft campaign data are removed.

We conduct additional analysis to examine the influence of spatial versus temporal sampling biases (**Fig.S3**). When co-sampled with observations only in space with monthly mean ozone fields archived from the model for each April-May, AM3_BASE simulates an ozone increase (0.78 ± 0.34 ppbv yr⁻¹ for 1995-2008) similar to the trend when co-sampled both in space and time. The estimates of regional ozone median with either of the two co-sampling approaches are biased low in the earlier years as compared to the ‘true median’, indicating that year-to-year variability in the ozone spatial pattern and changes in measurement locations influence the calculated ozone trends. Although the ozone trends inferred for 1984-2008 and 1995-2008 are most influenced by spatial sampling biases, temporal sampling deficiencies also contribute (orange vs. green symbols in **Fig.S3**). Since free-running climate models are not expected to

capture observed interannual meteorology, they are unlikely to reproduce the ozone trend found by *Cooper et al.* [2010], even co-sampling in space from monthly means.

3.2 The 1984-2014 and 1995-2014 trends

We extend the analysis of *Cooper et al.* [2010] using additional data to 2014 from ozonesondes, lidar and aircraft data, including ozone measurements from the NASA Ames Alpha Jet Atmospheric eXperiment [Yates et al., 2013]. Sample size for 2009-2014 (approximately 1900-4800 per year; Fig.S4) is similar to the earlier datasets. Supplementary **Table S1** summarizes the ozone trends inferred for several time periods, including the 1984-2011 and 1995-2011 periods analyzed by *Cooper et al.* [2012] and *Parrish et al.* [2014]. Extending the analysis to 1995-2014 (1984-2014), we find an ozone trend of 0.31 ± 0.21 (0.42 ± 0.17) ppbv year⁻¹ from observations and 0.36 ± 0.18 (0.27 ± 0.10) ppbv year⁻¹ in AM3_BASE ‘true’ median (**Fig.1a and Table S1**). The agreement of the 1995-2014 trends is probably due to meteorological variations averaging out to a greater extent over the longer time period. Since observed ozone levels have remained stable in the most recent six years, the trend for 1995-2014 is only ~50% as large as for 1995-2008. In contrast, the model ‘true’ trend changes from an insignificant increase of 0.25 ± 0.32 ppbv year⁻¹ for 1995-2008 to a significant increase of 0.36 ± 0.18 ppbv year⁻¹ for 1995-2014. When sampled sparsely, ozone in AM3_BASE shows a leveling-off trend as in the observations (**Fig.1a**), although the growth rate for 1995-2014 (0.53 ± 0.21 ppbv year⁻¹; **Table S1**) is greater than observed. This likely reflects the combined influences from sparse sampling and emission uncertainty in the model (Section 4).

Comparison with the model-derived fields therefore suggests that the extensive statistical

sensitivity analysis of *Cooper et al.* [2010] was not sufficient to control for the influence of meteorological variability and sampling biases in their data since that information was not captured by the data. However, if we can assume that the meteorologically-driven ozone variability in the model forced with reanalysis winds is approximately correct, at least in magnitude, then the differences between the model ‘true’ median points and co-sampled with observations can be used as a measure of the “data representativeness uncertainty” (See Supplementary Methods). This representativeness uncertainty can be added to the statistical uncertainty on the trend derived from observations, so that the estimated trend for 1995-2008 becomes 0.65 ± 0.57 ppbv year⁻¹, which overlaps with the ‘true median’ trend of 0.25 ± 0.32 ppbv year⁻¹ derived from AM3_BASE. When our (model-based) estimate of representativeness uncertainty is included, the new trend for 1995-2014 becomes 0.31 ± 0.32 ppbv year⁻¹.

4. Roles of anthropogenic emission trends versus internal climate variability

[Figure 3 about here]

We next examine the roles of anthropogenic emission trends versus internal climate variability, based on AM3 ‘true median’ results (**Fig.3**). To establish credibility in the model attribution of the ozone response to rising Asian emissions, we compare trends in tropospheric NO₂ columns over Eastern China as observed by GOME/SCIAMACHY and simulated by AM3_BASE (**Fig.3a**). Our emission inventory (see Methods) captures the increasing NO₂ column abundances over eastern China during 1996-2011 as retrieved from the satellite instruments. The decreased NO₂ rate of change from SCIAMACHY during 2008-2010 likely reflects the influence of the Chinese economic downturn [*Lin and McElroy*, 2011], which is not

represented in the emission inventory. The rate of NO_x emission increase during the most recent decade is larger than during 1980-2000. A number of studies have documented the doubling of Chinese NO_x emissions over the past decade [Zhang *et al.*, 2009; Hilboll *et al.*, 2013; Kurokawa *et al.*, 2013]. In contrast, US NO_x emissions have declined by ~50% in the 2000s [Kim *et al.*, 2006; Lamsal *et al.*, 2015; Tong *et al.*, 2015].

Ozone over western North America simulated with constant emissions has declined overall from the early 1980s to the mid-1990s and rebounded afterwards (**Fig.3b**), coinciding with observed decadal variability of lower stratospheric ozone burdens in the 35-year Canadian ozonesonde records [Tarasick *et al.*, 2005; Oltmans *et al.*, 2013; Lin *et al.*, 2015]. Year-to-year fluctuations are strongly correlated with the stratospheric influence ($r^2=0.65$), which can confound the attribution of ozone changes in short observation records to anthropogenic emission trends, as illustrated for the 1992-2012 period. Over the entire 1980-2014 period, we find no significant trend in the stratospheric influence. Over 1992-2012, however, we find an increase of 0.36 ± 0.23 ppbv year⁻¹ in O₃Strat and 0.21 ± 0.14 ppbv year⁻¹ in total mid-tropospheric ozone in AM3 with constant emissions (**Fig.3b**). This increase is attributed in part to anomalies driven by the 1991 Mt. Pinatubo volcanic eruption and by recent La Niña events [Lin *et al.*, 2015].

With both emissions and meteorology varying, AM3_BASE simulates an ozone increase of 0.38 ± 0.18 ppbv year⁻¹ over 1992-2012 and 0.22 ± 0.08 ppbv year⁻¹ over the entire 1980-2014 period (**Fig.3c**). With North American anthropogenic emissions shut off, AM3 gives an increase of ‘background’ ozone of 0.46 ± 0.15 ppbv year⁻¹ over 1992-2012 and 0.29 ± 0.07 ppbv year⁻¹ over

1980-2014. We examine the trends under conditions when simulated ozone is most influenced by direct transport from Asia, as diagnosed by East Asian CO tracer exceeding the 67th percentile for each April-May (see Methods in *Lin et al.*, 2014). Simulated ozone background under strong transport from Asia increases at a greater rate of 0.55 ± 0.14 ppbv year⁻¹ over 1992-2012, consistent with the stronger trend found by *Cooper et al.* [2010] when subsampling the observational datasets for Asian influence, although we note that ozone from stratospheric intrusions can also be mixed into Asian pollution plumes [e.g. *Cooper et al.*, 2004; *Lin et al.*, 2012b].

Finally, we evaluate decadal mean changes (2005-2014 minus 1980-1989) to differentiate a significant ozone change signal forced by emissions from one due to large interannual variability in meteorology (**vertical bars in Fig.3b**). AM3 O₃Strat shows a positive but insignificant increase (1.9 ± 2.9 ppbv) from 1980-1989 to 2005-2014, which possibly offsets other circulation-driven ozone decreases in the model, leading to no change in total mean ozone levels in FIXEMIS. The decadal mean rose 5.9 ± 2.1 ppbv under BASE with emissions varying and 7.6 ± 2.0 ppbv under Background with North American emissions shut off. The increase in mean ‘background’ ozone over western North America from the 1980s to the most recent decade is well above the meteorologically driven ozone change under FIXEMIS and above the 95% confidence intervals, indicating the key roles of rising Asian emissions and global methane.

5. Conclusions and implications for additional ozone measurements

We have investigated the factors driving variability and long-term changes of springtime ozone in the free troposphere over western North America during the past 35 years. The model

‘true median’ indicates that mean springtime ozone levels over western North America in the most recent decade has increased by 5.9 ± 2.1 ppbv compared to the 1980s (**Fig.3**). With North American anthropogenic emissions shut off, the decadal mean ‘background’ trend is 7.6 ± 2.0 ppbv. These increases, attributed in part to rising Asian ozone precursor emissions and global methane, are consistent with rising baseline ozone observed at northern mid-latitude remote sites [e.g. Parrish *et al.*, 2012]. Nevertheless, the substantial variability in ozone resulting from meteorologically driven, sometimes offsetting, factors poses a challenge to the detection of ozone changes from short (and sparse) observation records and the attribution of these changes to anthropogenic emission trends, as illustrated for the 1995-2008 and 1992-2012 periods. Meteorologically-driven ozone variability has large enough amplitude to render the strong, apparently very robust trend for 1995-2008 found in the model co-sampled with sparse observations non-significant when sampling biases are fully considered (**Fig.1**).

Multi-decadal hindcast simulations driven by observed meteorology can aid the interpretation of tropospheric ozone observations, especially with regard to data representativeness and attribution. Our study provides new context for the interpretation of trends in chemical constituents from observations and models being conducted for the International Global Atmospheric Chemistry (IGAC) Project’s Tropospheric Ozone Assessment Report (TOAR) and Chemistry-Climate Model Initiative (CCMI). We show how apparent conflicts between observations and model results can be reconciled by making an apples-to-apples comparison. Considering the complications introduced by internal climate “noise” in observational records and free-running chemistry-climate models, we instead recommend comparing observed trends

in chemical constituents with models driven by observed meteorology.

Advancing knowledge on ‘background’ ozone can provide valuable ozone source attribution information to air quality control managers as they develop ozone abatement strategies under a lowered U.S. ozone air quality standard [US EPA 2014; *Fiore et al.*, 2014; *Cooper et al.*, 2015], particularly for high-elevation western U.S. regions, where deep mixed layers facilitate the transport of free tropospheric ozone to the surface. Accurate quantification of ‘background’ ozone requires enhanced observations at a spatial density and temporal frequency adequate for evaluating and improving the models. The present-day ozone sampling frequency and distribution does not capture the full interannual and spatial variability of ozone across western North America, especially on time scales as short as 14 years (1995-2008). While satellite retrievals can provide a large-scale view of mean mid-tropospheric ozone distributions [e.g. *Verstraeten et al.*, 2015], the current record length (<10 years) is too short for robust trend analysis. Accurate detection of changes in springtime ozone means and extremes over the western U.S. [*Lin et al.*, 2015] will require improvements in the current observation network.

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Acknowledgments

This work is supported by funding from NASA Atmospheric Composition-Aura Science Team (NNH13ZDA001N-AURAST) to M.-Y. Lin and O.R. Cooper. We acknowledge the NOAA/GMD/CCGG Aircraft Project (PI: Colm Sweeney) and support of NASA Ames Research Center partnered with H211 L. L. C. (Alpha Jet Atmospheric eXperiment) for their role in collecting ozone samples. We are grateful to V. Naik, F. Paulot, and D.D. Parrish for reviewing an earlier version of the paper. Model simulations are archived at GFDL and are available to the public upon request to M.-Y. Lin.

Figure Captions

Figure 1 | Representativeness of ozone measurements constructed from various platforms.

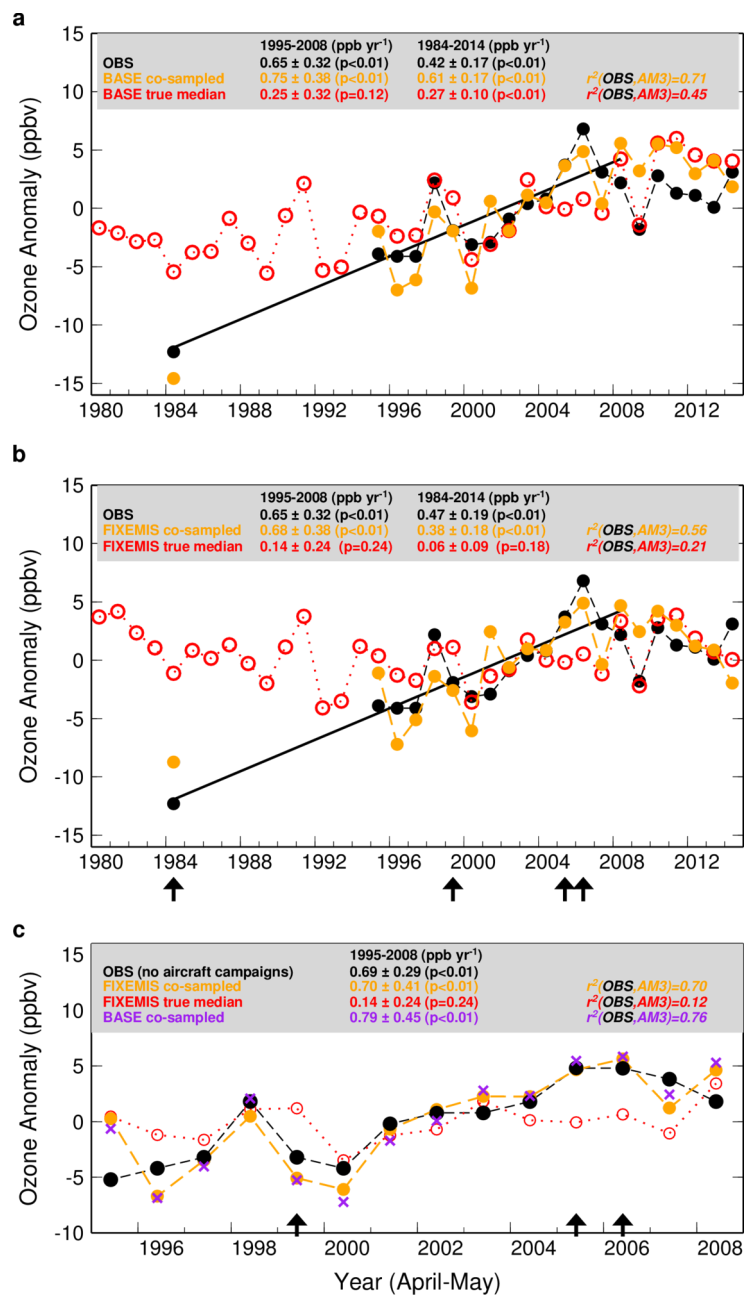
(a) Anomalies (relative to 1995-2010) in the median values of April-May ozone mixing ratios 3-

8 km above western North America for 1980 to 2014: from a composite of available *in-situ* measurements (black); from AM3_BASE co-sampled in space and time with observations (orange); and from the model ‘true median’ of continuous daily and spatial sampling over the entire domain (32-50N, 128-104W; red). **(b)** Same as (a) but for AM3_FIXEMIS. **(c)** For the 1995-2008 period with aircraft campaign data removed. The linear trends, the 95% confidence intervals, p-values and correlations (r^2) between observations and simulations are shown. Arrows at the bottom of the graph denote the years when observational estimates of regional median ozone are biased by sampling artifacts (**Fig.2**).

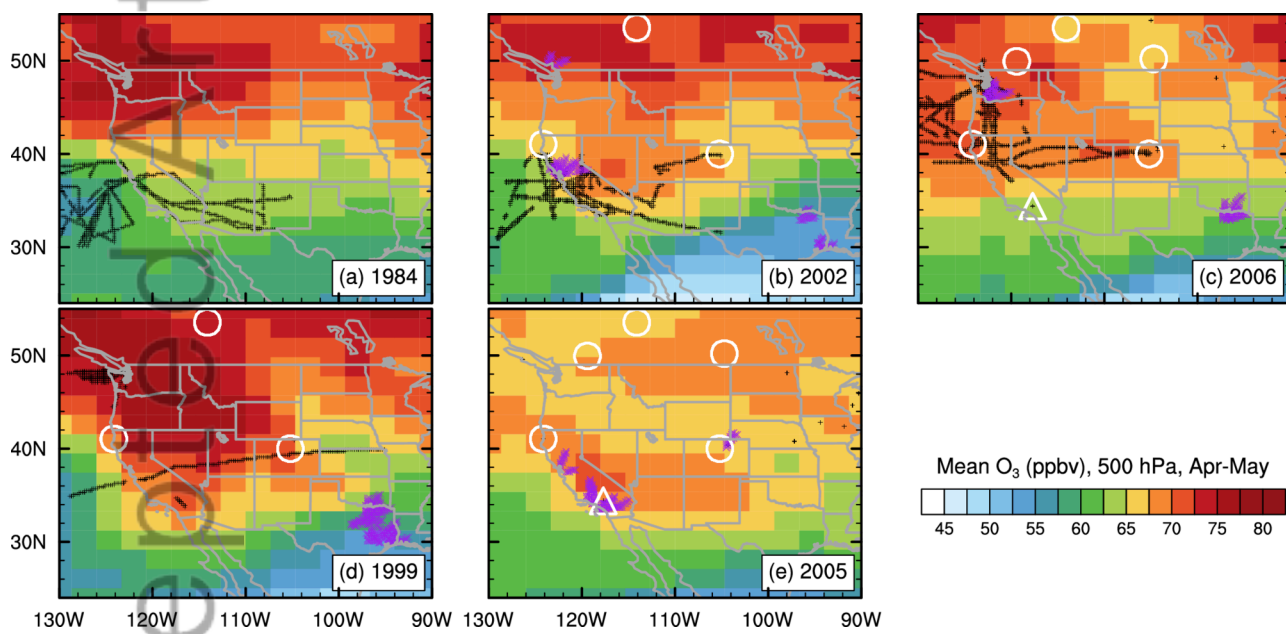
Figure 2 | Influence of meteorological variability and sparse in-situ sampling. Sampling locations superimposed on the mid-tropospheric ozone maps from AM3_FIXEMIS. Measurement platforms are labeled: aircraft field campaigns (black ++), MOZAIC commercial aircraft between 3-8 km altitude (purple x), ozonesondes (O), and Table Mountain lidar (Δ).

Figure 3 | Roles of rising Asian emissions versus internal climate variability. **(a)** Mean annual tropospheric NO₂ columns over eastern China normalized to the year 2000 from GOME (1996-2002, open circles) and SCIAMACHY (2003-2011, closed circles) measurements and from AM3_BASE sampled at satellite overpass time (10:30AM) (purple lines). Similar trends in NO_x emissions (purple circles) indicate that the influence of NO_x lifetime is of minor importance on the trend in morning NO₂ columns. Satellite NO₂ data are downloaded from www.temis.nl, with retrieval technique described in [Boersma *et al.*, 2004]. **(b)** Anomalies in the ‘true median’

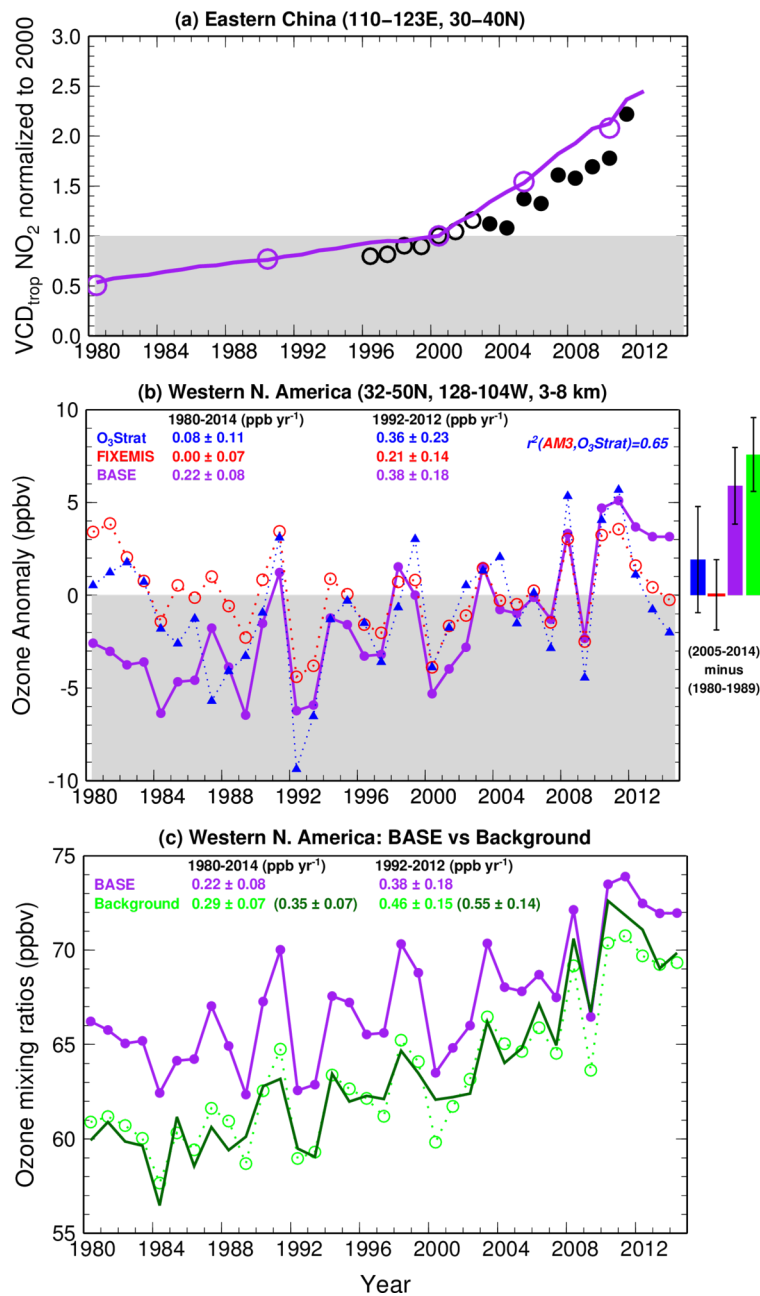
values of April-May ozone 3-8 km above western North America simulated by AM3_BASE (purple), FIXEMIS (red) and stratospheric ozone tracer (blue; O₃Strat). (c) Median ozone in BASE (purple) versus Background with North American emissions shut off (green). The darkgreen line denotes Background under conditions with strong transport from East Asia (trends shown in parentheses). Vertical bars show decadal mean changes (with the 95% confidence intervals) between 1980-1989 and 2005-2014.



figure_1



figure_2



figure_3